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Preparation and non-destructive characterization of $YBa_2Cu_3O_{7-\delta}$ films for microwave applications

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Abstract

We have deposited high-quality $YBa_2Cu_3O_{7-\delta}$ films with a thickness between 200 nm and 500 nm and a size of 1 cm^2 on YSZ, MgO and $NdGaO_3$ substrates. Shielding measurements show that critical temperatures T_c between 89 K and 93 K and transition widths ΔT_c between 0.7 K and 0.2 K can be obtained reproducibly on all of these substrates. A contactless inductive j_c measurement technique, based on the detection of the 3rd harmonic, has given critical current densities up to (5.1 ± 0.5) MA/cm^2 at 77 K. For the best films, values of the surface resistance R_s at 87 GHz and 77 K of (16 ± 3) mΩ have been derived from measurements in a copper host cavity. Furthermore, these films show a significant reduction of the residual microwave losses at temperatures below 60 K, leading to R_s values below 3 mΩ at 4.2 K. From such high-quality $YBa_2Cu_3O_{7-\delta}$ films on MgO, we have started to fabricate and optimize the design of microwave devices by use of standard patterning techniques. As an example, a narrow nonlinear coplanar-line device is presented. We have observed "fast" and "slow" nonlinear response, which limits the output power.

1. Introduction

Due to the specific features of HTS oxides and the state of existing technology, planar passive microwave devices are among the first applications of these superconductors [1]. In this paper, we demonstrate that planar on-axis DC-sputtering from stoichiometric targets can be used to produce high-quality $YBa_2Cu_3O_{7-\delta}$ films for microwave devices. Furthermore, we present the properties of a narrow coplanar-line microwave device, which acts as a power limiter and which is also useful for the investigation of the nonlinear response of HTS.

2. On-axis sputter process

Among the epitaxial film growth techniques, planar on-axis DC-sputtering under an oxygen pressure of a few mbar needs only a rather simple and cheap apparatus to produce smooth high-quality $YBa_2Cu_3O_{7-\delta}$ films. The special feature of this technique, which has been introduced for HTS films by U. Poppe from Forschungszentrum Jülich (Germany) [2], is the relatively high oxygen pressure during the deposition. This offers a lot of oxygen to the growing film and minimizes resputtering effects [3]. The resputtering is supressed, because high-energy negatively-charged particles with an energy up to a few hundred eV are slowed down by inelastic collisions. However, the short mean free path of the particles of a few μm causes small deposition rates of only 50-80 nm/h. Compared to sputter processes with Argon, the use of a few mbar of pure oxygen is advantageous, because this leads after a short sputtering time to very stable and reproducible sputtering conditions [3]. As a consequence, this special on-axis sputter process is relativly slow but well controllable.

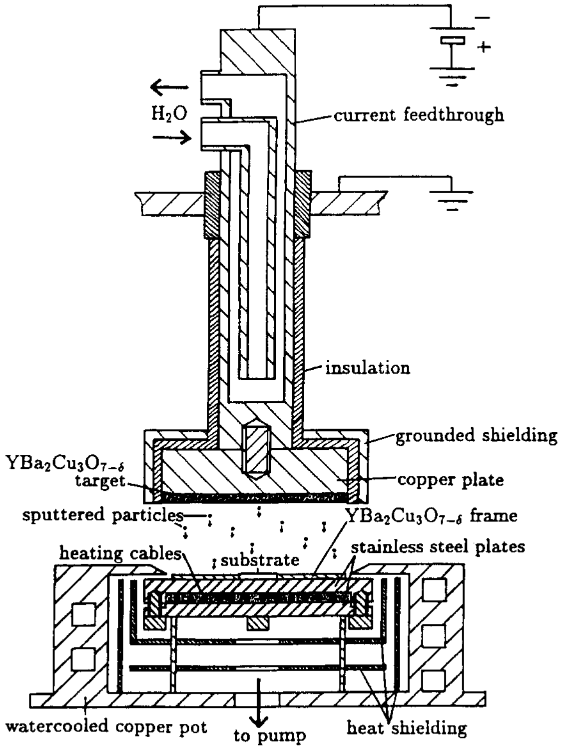

Figure 1: On-axis DC-sputtering system.

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Fig.1 shows a schematic picture of the sputtering system. It consists of two main components – the cathode and the substrate heater. The stoichiometric $YBa_2Cu_3O_{7-\delta}$ target with a diameter of 35 mm is bonded to a watercooled current feedthrough. The feedthrough is surrounded by insulation and partially also by grounded shielding. This leads to a stable stationary gas discharge between the target and the substrate heater. The substrate heater consists of a bifilar spiral of heating cables between two stainless steel plates. For better thermal contact, the spiral and a thermocouple have been embedded in BN powder and is sintered at about 900° C for 6 h. The heater is surrounded by a system of heat- shields, openings and a watercooled copper pot in order to improve the temperature homogeneity of the heater and to protect the vacuum chamber from too much heat. The substrate is placed on top of the heater and is surrounded by a silver frame, which is coated with $YBa_2Cu_3O_{7-\delta}$[4]. This frame acts also as a heat shielding, avoids contamination from the heater plates and homogenizes the oxygen flow at the edge of the substrate. Oxygen flow during the deposition is achieved by pumping the system directly below the heater and using a regulated gas inlet somewhere in the chamber.

The deposition rate is mainly dependent on the sputter current and the oxygen pressure and flow. For our geometry, we have obtained sufficient deposition with an oxygen pressure of (3.85 ± 0.01) mbar, a flow of more than 21 l/h and a current of 150 mA. With this set of parameters, we received in 4 h deposition time film thicknesses of about 300 nm. We have kept this thickness for the whole optimization.

The target stoichiometry has a strong influence on the composition of the the HTS films and therefore on their superconducting, morphological and structural properties [5]. Furthermore, we have observed that not only the average stoichiometry but also the phase purity and grain structure of the target influence the film quality. The average target stoichiometry and homogeneity have been controlled by EDX. Our results suggest that reproducible film preparation by on-axis DC-sputtering can only be achieved with stoichiometric targets which have high phase purity and uniform grain structure.

The substrates have also a strong influence on the film quality. We have observed by light microscopy and SEM different outgrowth densities σ with different average diameters D for optimized films on YSZ ($\sigma \approx 340000\,\mathrm{mm}^{-2}$,D $\approx$ 300 nm), on MgO ($\sigma \approx 17000\,\mathrm{mm}^{-2}$,D $\approx$ 700 nm) and on $NdGaO_3$ ($\sigma \approx 45000\,\mathrm{mm}^{-2}$,D $\approx$ 400 nm). In addition, we have found a change in the average T_c and ΔT_c from $\overline{T_c} = 90.8$ K and $\overline{\Delta T_c} = 0.7$ K for optimized films on MgO to $\overline{T_c} = 92$ K and $\overline{\Delta T_c} = 0.5$ K for films on $NdGaO_3$. As MgO allows growth of high-quality films and provides in contrast to YSZ and $NdGaO_3$ low dielectric losses, we have used it as our standard substrate.

Another critical parameter of the planar on-axis DC-suttering process is the spacing between the target and the substrate d_{TS}. In general, the substrate has to be placed within the boundary region of the oxygen plasma to receive high-quality films [3]. We found that the superconducting film properties improve slightly for smaller d_{TS}. However, their surface shows more outgrowth. As a preliminary compromise, we have fixed d_{TS} to 9 mm.

The main preparation parameter which has to be optimized for a given target, substrate and d_{TS} is of course the combination of temperature and oxygen pressure during deposition [6, 7] and cooling [8, 9]. For a fixed oxygen pressure of 3.85 mbar, we have varied the film temperature systematically. We have observed for the heater temperature, measured with the thermocouple, a nearly 80° C wide window around 835° C for film growth on MgO and an at least 60° C around 935° C for $NdGaO_3$, respectively. For too low growth temperature, we observed incomplete epitaxial growth, yielding granularity and non-uniform orientation of the films, combined with poor superconducting properties. Too high growth temperature resulted in very poor film quality, caused by the decomposition of the 1:2:3 phase [6, 7]. So far, we have cooled our films slowly from deposition temperature to room temperature with a rate of 10 – 50° C/min in oxygen atmosphere.

3. Film characteristics

We have measured the superconducting properties of the $YBa_2Cu_3O_{7-\delta}$ films exclusively by nondestructive methods in order to fabricate devices with these films. Shielding measurements show that critical temperatures T_c between 89 K and 93 K and transition widths ΔT_c between 0.7 K and 0.2 K can be obtained reproducibly on YSZ, MgO and $NdGaO_3$. For the latter both, a contactless inductive j_c measurement technique based on the detection of the third harmonic [10, 11] has given critical current densities up to $(5.1 \pm 0.1)\,\mathrm{MA/cm}^2$ at 77 K. In Fig.3 the temperature dependent surface resistance $R_s(T)$ of selected films on MgO and $NdGaO_3$ is presented. The films have been measured in a copper host cavity at 87 GHz. These films provide T_c values between 91 K and 93 K and show a sharp drop of R_s to values of about 20 mΩ at 77 K. Similar to other sputtered high-quality films [9] a significant reduction of R_s has been observed below 60 K. This can be interpreted as a sign for a two gap system of superconducting planes and chains as introduced recently by Kresin and Wolf [12]. The anomalous behavior of R_s at low temperatures ($<$ 30 K) will not be discussed in detail in this paper. However, it is worthwhile to mention that there seems to be a correlation between these fluctuations and the also strongly varying normal state conductivity of these films. Within the mentioned model, these features are considered as a result of the chain influence [8, 12]. We have measured 22 films deposited with optimized parameters on MgO and $NdGaO_3$ at 87 GHz. 73 % of these films have R_s values between 18 mΩ and 26 mΩ at 77 K and nearly all of them show R_s values below 18 mΩ at 4.2 K. For the best films R_s values of (16 ± 3) mΩ at 77 K and below 3 mΩ at 4.2 K have been measured at 87 GHz.

Because of the above mentioned superconducting properties, the thickness of about 300 nm and its variation of less than ±3 %, our films are well suited for microwave devices. For some special devices, films with similar properties and thicknesses down to 200 nm and up to 500 nm respectively have been produced as well. In addition, a smooth film surface is desired as roughness, outgrowth and troughs limit the performance of multilayer strutures and also the patterning facilities. AFM measurements and SEM pictures show that our films are smooth except for the already mentioned outgrowths and troughs.

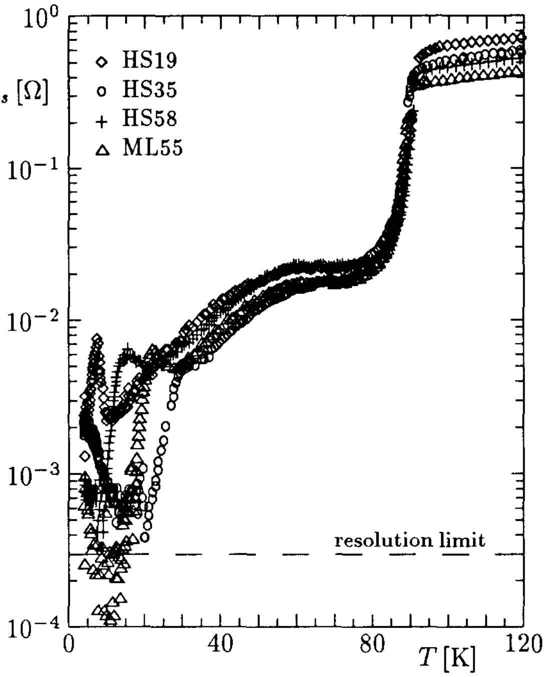

Figure 3: $R_s(T, 87\,\mathrm{GHz})$ of films on MgO and $NdGaO_3$.

4. Device production and characterization

We have produced microwave devices from the characterized $YBa_2Cu_3O_{7-\delta}$ films. The main components of modern microwave devices can be devided into three groups [13]:

1. radiating linear microwave devices
   (e.g. antennas)
2. non-radiating linear devices
   (e.g. filters and delay lines)
3. non-radiating nonlinear devices
   (e.g. limiters, switches and mixers)

The first two groups profit from the extremely low losses of the $YBa_2Cu_3O_{7-\delta}$ films. Therefore, the dimensions of these devices can be reduced. As the degree of miniaturization depends on the dielectric constant ϵ_r of the substrate a relatively high $\epsilon_r \geq 20$ and also low dielectric loss tangent $\tan\delta \leq 10^{-4}$ is desired [13]. As a consequence, MgO and $NdGaO_3$ are not suitable for such devices. Non-radiating nonlinear devices use a specific feature of HTS, the fast changes of microwave properties in dependence of external controlling fields. As an example, a narrow nonlinear coplanar-line device is presented in fig.4.

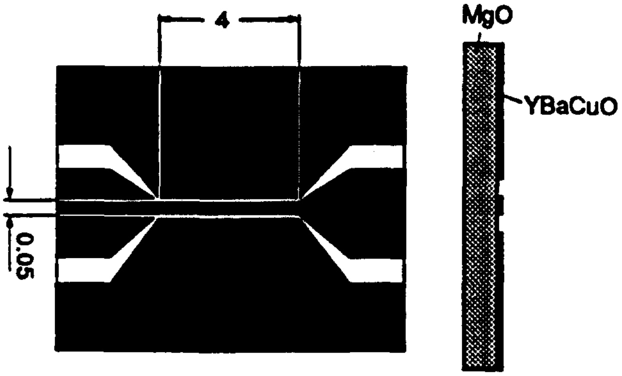

Figure 4: Narrow, nonlinear coplanar-line device

This device is based on films on MgO and has been alternatively patterned by wet and ion-beam etching. The films are spin-coated with approximately 1 μm of AZ photoresist. Using photomasks which limit the resolution to about 5μm, the coated films are exposed on a contact printer. Afterwards, the exposed resist is developed with standard AZ chemicals. Argon dry etching is done in a Kaufmann-type ion source with beam energies in the range of 0.5 keV to 1 keV. Alternatively for wet etching, we use saturated EDTA solution or dilute phoshoric acid. Finally, the samples are mounted into the fixtures and wired by silver loaded epoxy or ultrasonic bonding.

Two different types of nonlinear response can be

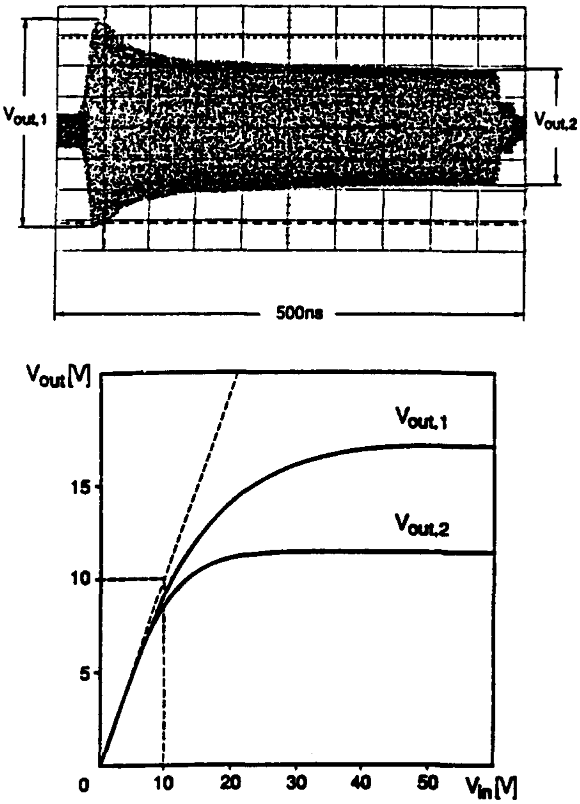

Figure 5: Observations of "fast" and "slow" nonlinear response measured with pulse-modulated microwave signals.

clearly observed from preliminary experiments with a pulse-modulated microwave signal [14]. The microwave signal, with a frequency of 2 GHz, was modulated with a video pulse which had a pulse length of 450 ns, a risetime of about 10 ns and a repition time of 6 μs. The resulting signal was then applied to the coplanar-line. A sampling oscillogram of the output (scale 2 V/div) signal at 77 K and the dependance of the output on the input signal, measured on a similar nonlinear microwave device, are presented in fig.5. The comparision of the output signal amplitudes $V_{out,1}$ of the leading and $V_{out,2}$ of the trailing edge with the amplitude of the input signal indicates that this device can be used as a limiter. Furthermore "fast" and "slow" nonlinearities can be clearly observed. The slow response can be identified as heating effects originating from the growth of "hot spots". The origin of the fast response is still a matter of controversy [14]. Additional measurements show that an efficient frequency conversion can be realized in short time-intervals, where thermal effects can be neglected. This has practical impacts, e.g. for the realization of samplers and correlators. Such devices are very promising, because the transition time is less than 25 ps.

5. Conclusion

We have prepared high-quality $YBa_2Cu_3O_{7-\delta}$ films by planar on-axis DC-sputtering from stoichiometric targets in order to fabricate superconducting devices. The influence of the preparation parameters oxygen flow, sputter current, target composition, substrate, spacing between target and substrate d_{TS}, oxygen pressure and growth temperature on the film quality has been investigated. In order to optimize both superconducting properties and surface morphology, we have already started to improve the target composition and to vary d_{TS} on a larger scale. We also plan to investigate the cool-down process more closely. In this context, the role of activated oxygen has to be studied, too. For the near future, we intend to use besides MgO also $LaAlO_3$ and $SrLaGa_3O_7$ substrates because of their promising dielectric and crystallographic properties. This will allow us to produce not only nonradiating nonlinear devices but also radiating and nonradiating linear microwave devices.

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